

Plutonium: An anthropogenic element under nuclear forensic investigations



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1. Introduction
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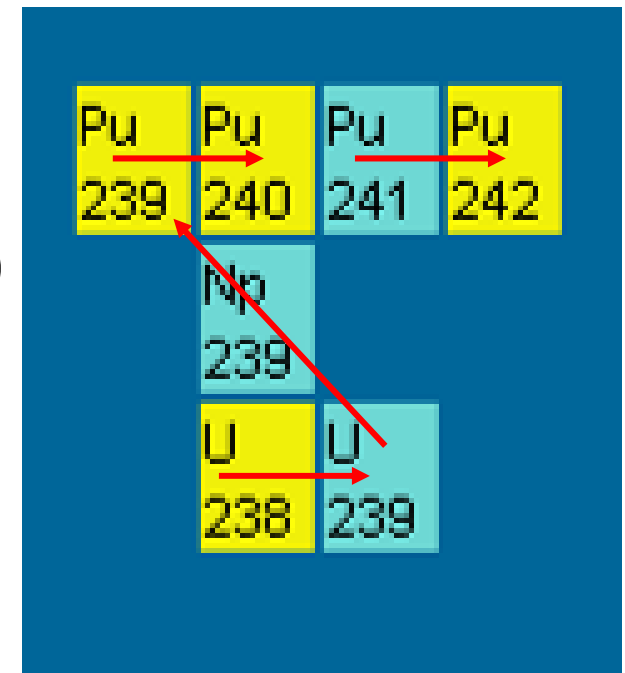
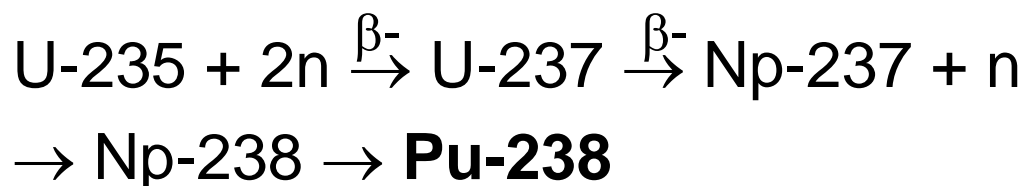
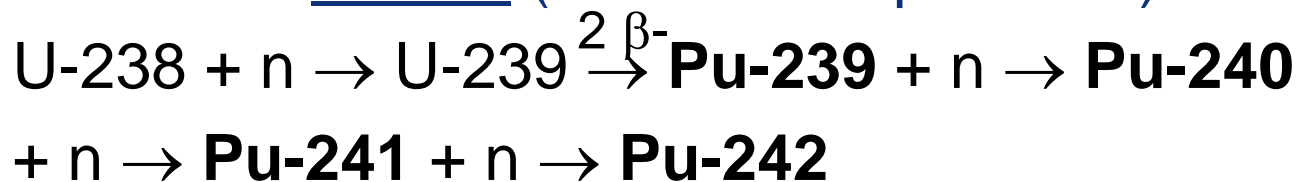


Plutonium is an anthropogenic, i.e. man-made element.

At the end of 1940 Pu-238 was first time synthesized:



In nuclear reactor (~1% Pu in spent fuel):



Main questions to be answered:

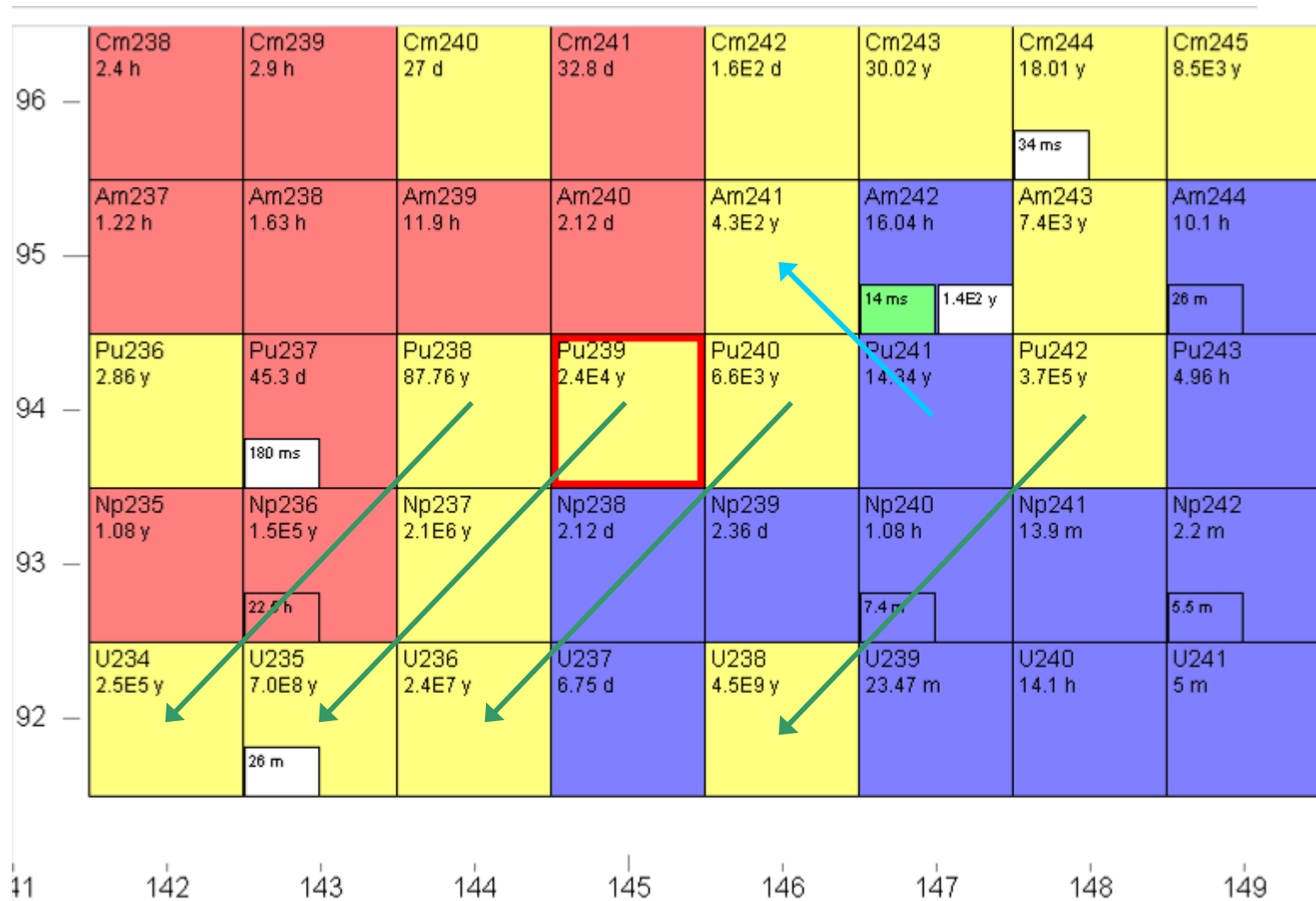
1. Intended use of the material
(nuclear weapons, fuel, scrap,...)

2. Origin

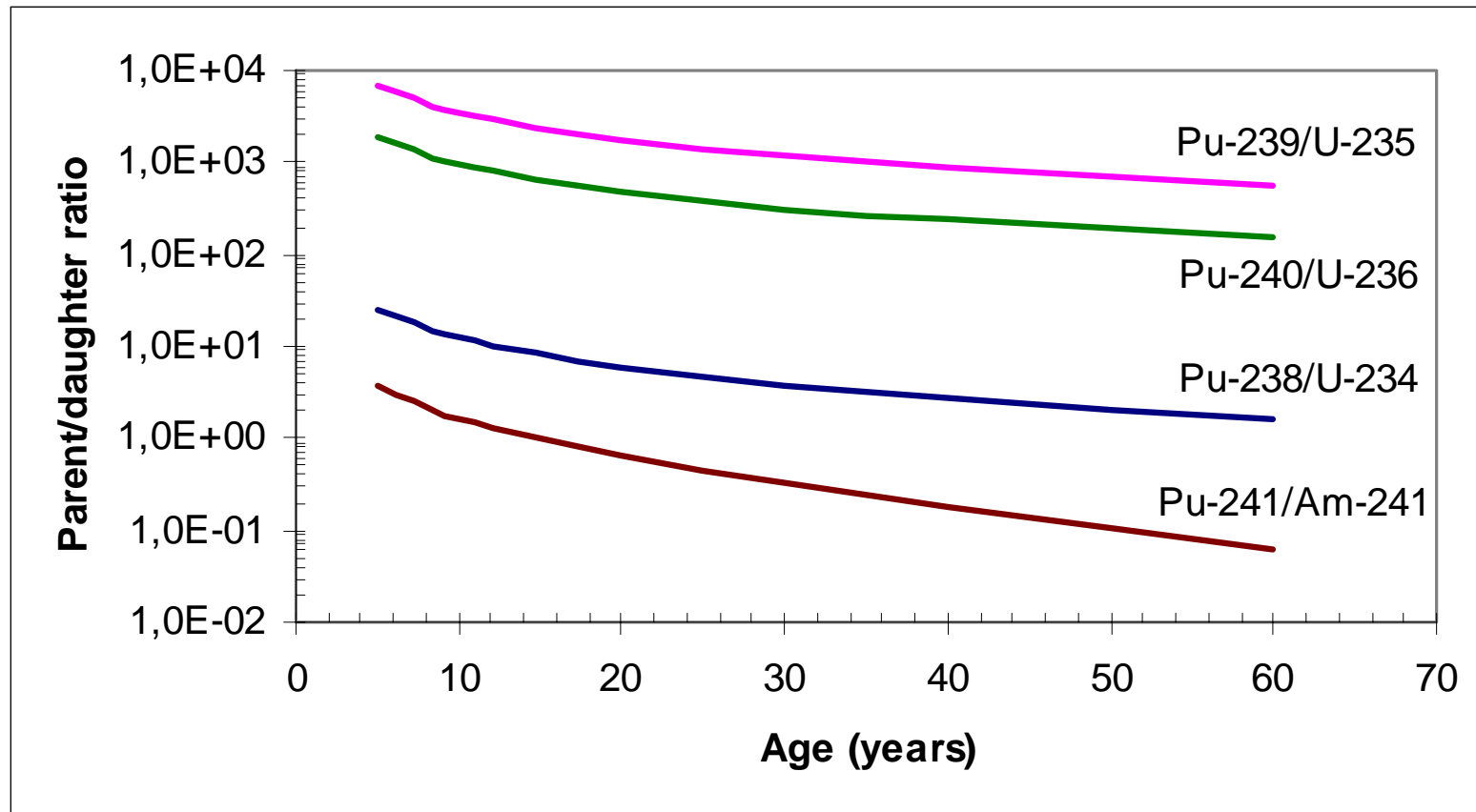
(place of production, process, place of
diversion, last legal owner,...)

Answers:

- isotopics
- Pu-content
- age
- impurities
- microstructure
- ...



Age = the time elapsed since the last chemical processing
(e.g. production, reprocessing, purification)



- Incomplete Pu/U/Am separation when reprocessing

⇒ U and Am remainings

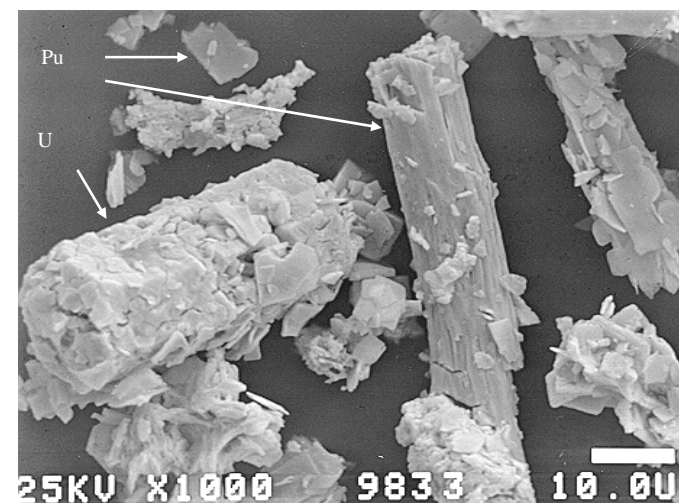
⇒ inconsistent results

Sample	$^{241}\text{Pu}/^{241}\text{Am}^*$	$^{238}\text{Pu}/^{234}\text{U}$	$^{239}\text{Pu}/^{235}\text{U}$	$^{240}\text{Pu}/^{236}\text{U}$
RR	6.1	2.35 ± 0.10	2.66 ± 0.26	2.29 ± 0.05
F19	20.6	1.18 ± 0.07	51.9 ± 3.8	19.9 ± 0.3

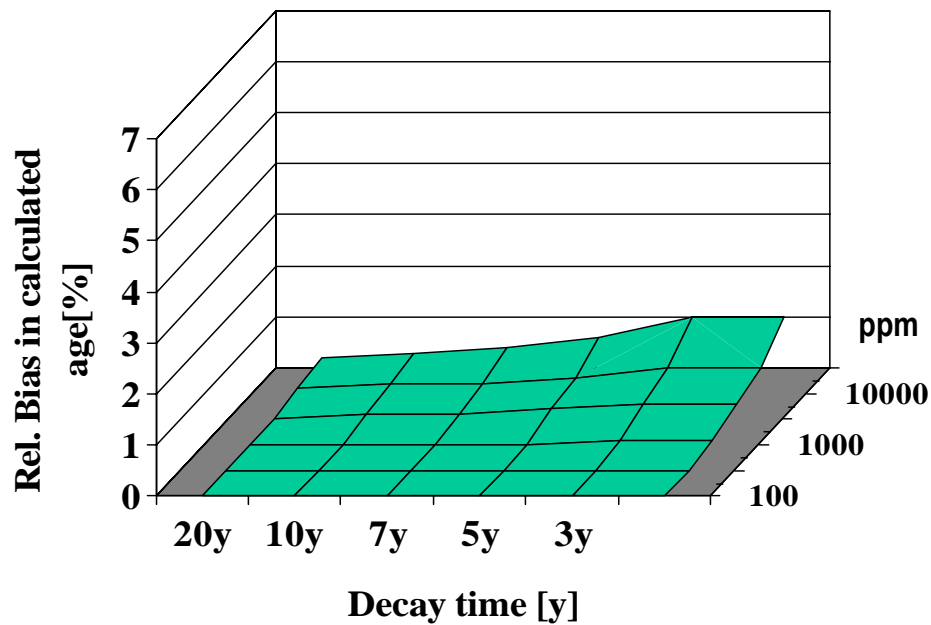
Age by SIMS in February 2000. * By gamma spectrometry.

- MOX

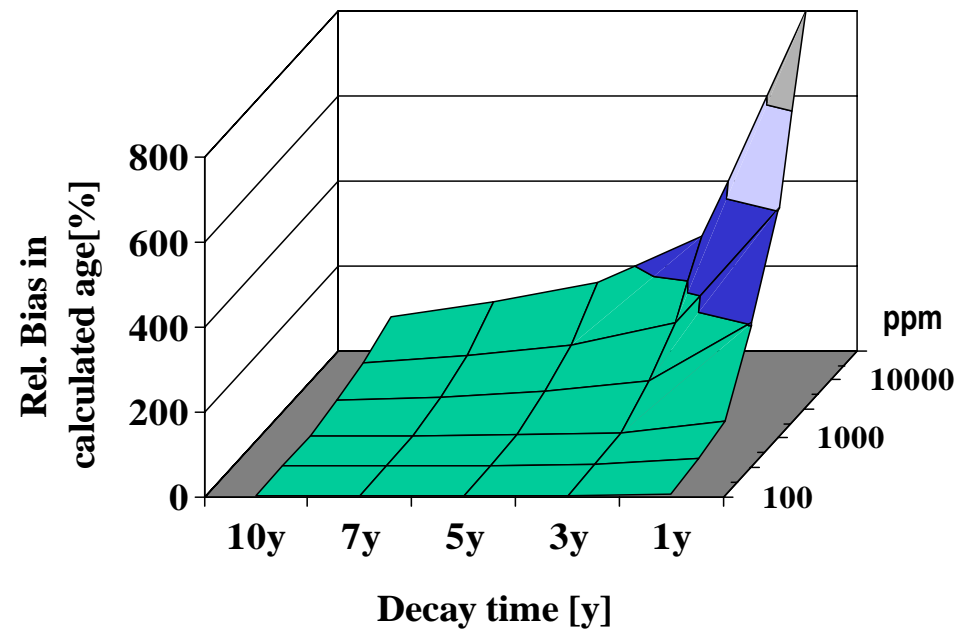
⇒ U interference



$^{238}\text{Pu} / ^{234}\text{U}$
reactor grade



$^{238}\text{Pu} / ^{234}\text{U}$
weapons grade



Weapons grade:

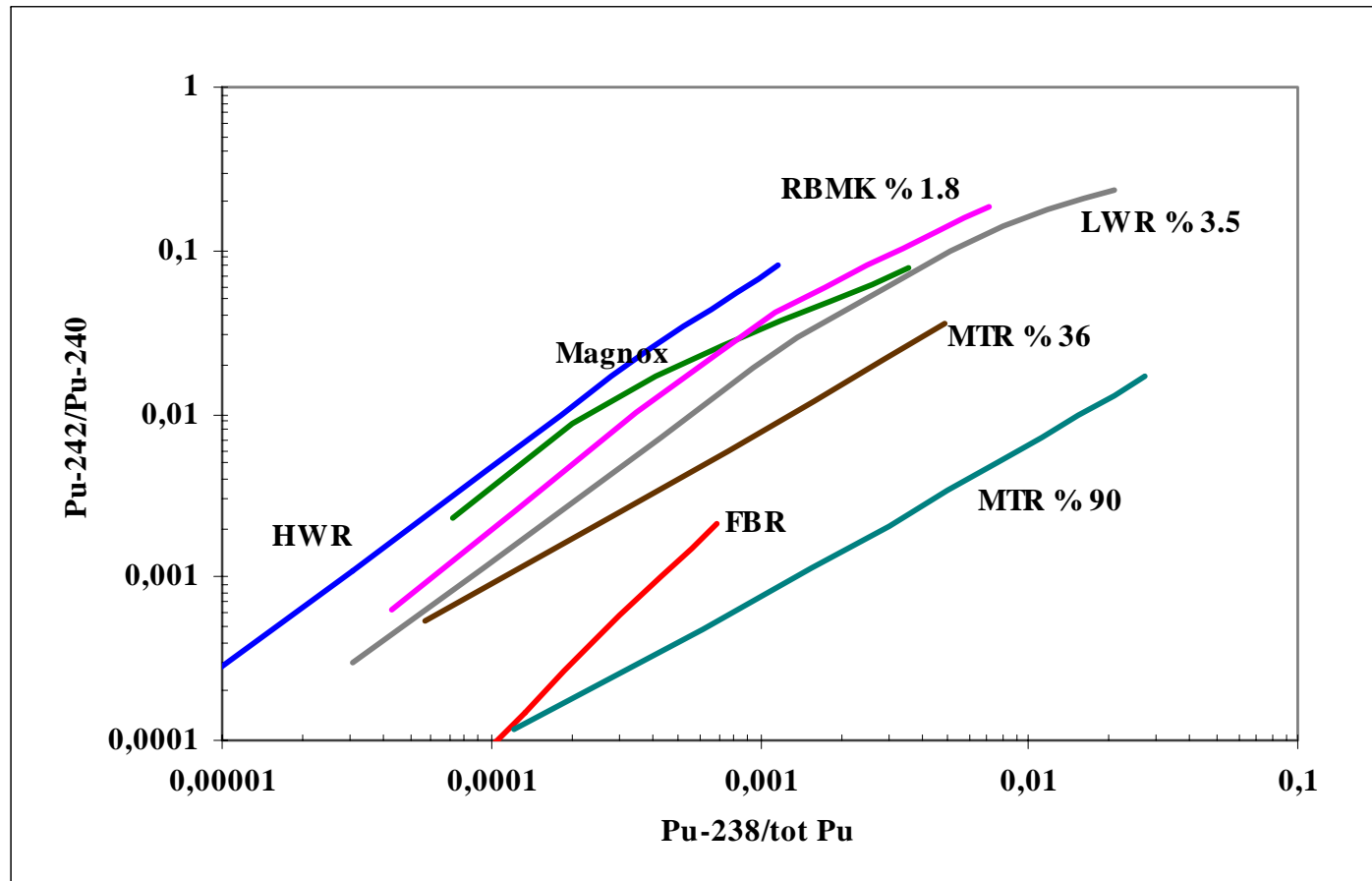
- Pu-240 < 7 %
- 240/239 < 0.07

Reactor grade:

- Pu-239 = 45-60%
- Pu-240 = 20-35 %
- 240/239 = 0.3-0.8



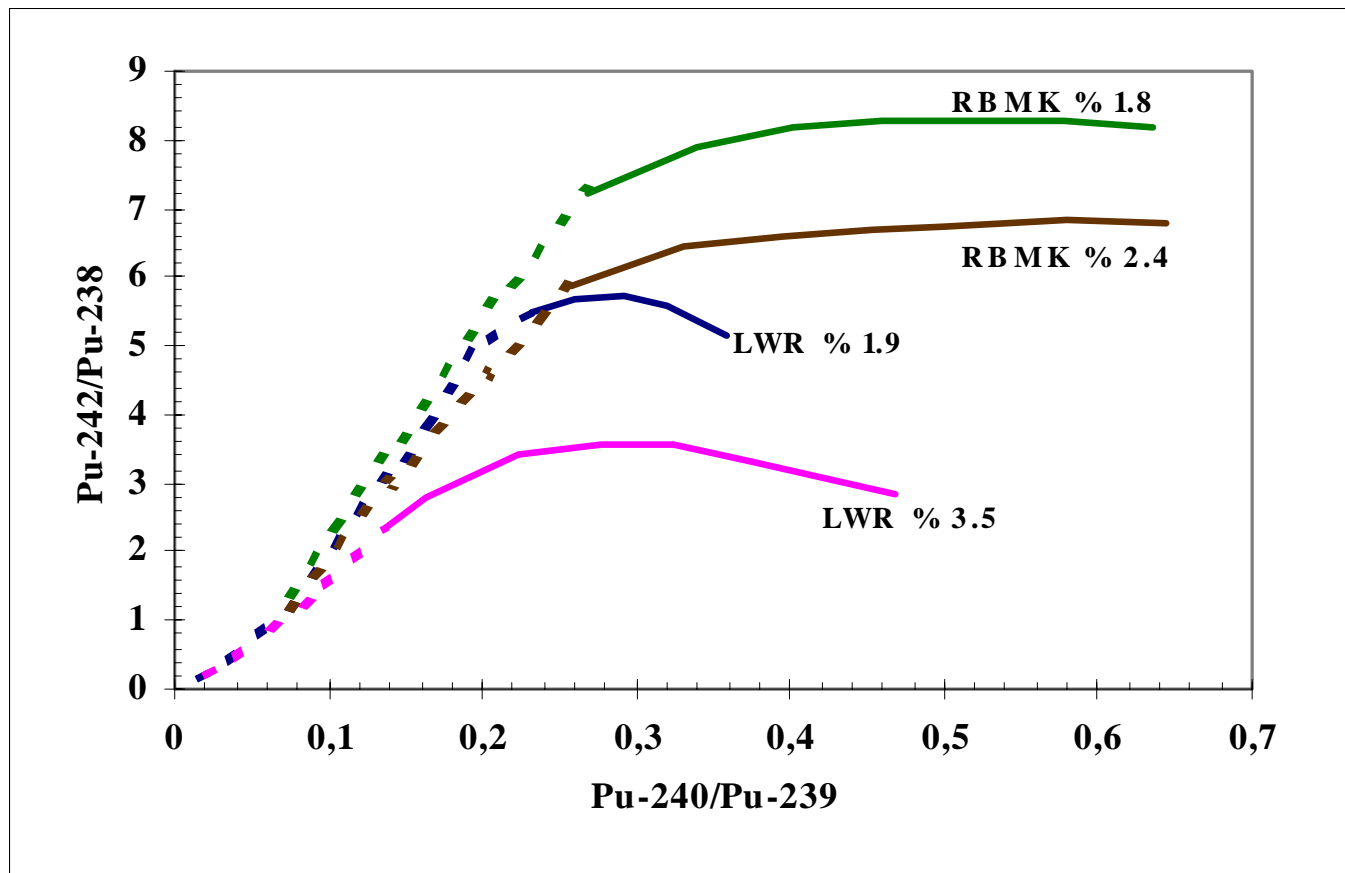
The isotopic composition of the reactor produced plutonium is depending on the initial U-235 enrichment, neutron spectrum (soft >< hard), flux and irradiation time.



↑
Softer the neutron spectrum – higher the $^{242}\text{Pu}/^{240}\text{Pu}$ ratio

Higher the initial ^{235}U enrichment – higher the ^{238}Pu abundance →

Difference between light-water and graphite-moderated reactors.



Case studies



56 g of powder containing Pu

Isotope/ element	w- %
^{238}Pu	0.067
^{239}Pu	99.751
^{240}Pu	0.181
Mercury	61
Antimony	11
Oxygen	6
Plutonium	10
Gallium	0.15 (4.6 a-%)
Iodine	1.6

$\text{Hg}_2\text{Sb}_2\text{O}_7$ = “Red mercury” !

Pu particle and powder agglomerate - electron probe



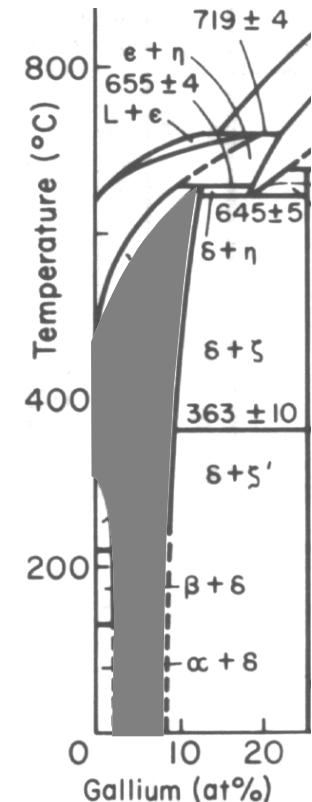
“Red mercury”:

- Anti-Radar-Shielding for aircraft's
- self-guided warheads
- nuclear bombs smaller and more efficient
- density 23 g/ccm

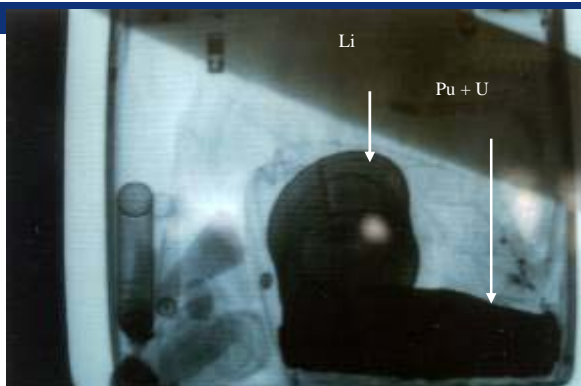
Compound with these characteristics has never been found !!

Plutonium:

- weapons-grade Pu
- Ga is used to stabilise the δ - phase of Pu



Example of the proliferation of classified knowledge !



Mixed plutonium and uranium oxides:

363 g Pu - 121 g U

Pu	238	239	240	241	242
w-%	0.17	87.58	10.78	0.81	0.66

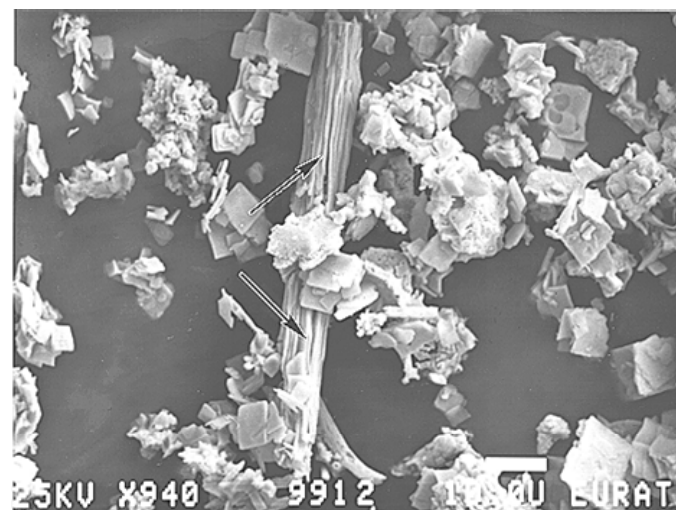
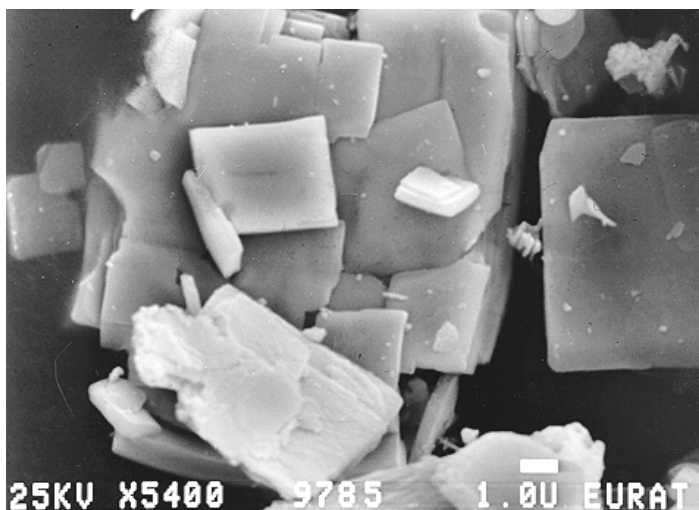
U	234	235	236	238
w-%	0.020	1.60	0.048	98.35

201 g of Li-metal enriched to 89.4 %
in ^6Li .

Powder consists of two different Pu particle types:

1) platelets of PuO_2

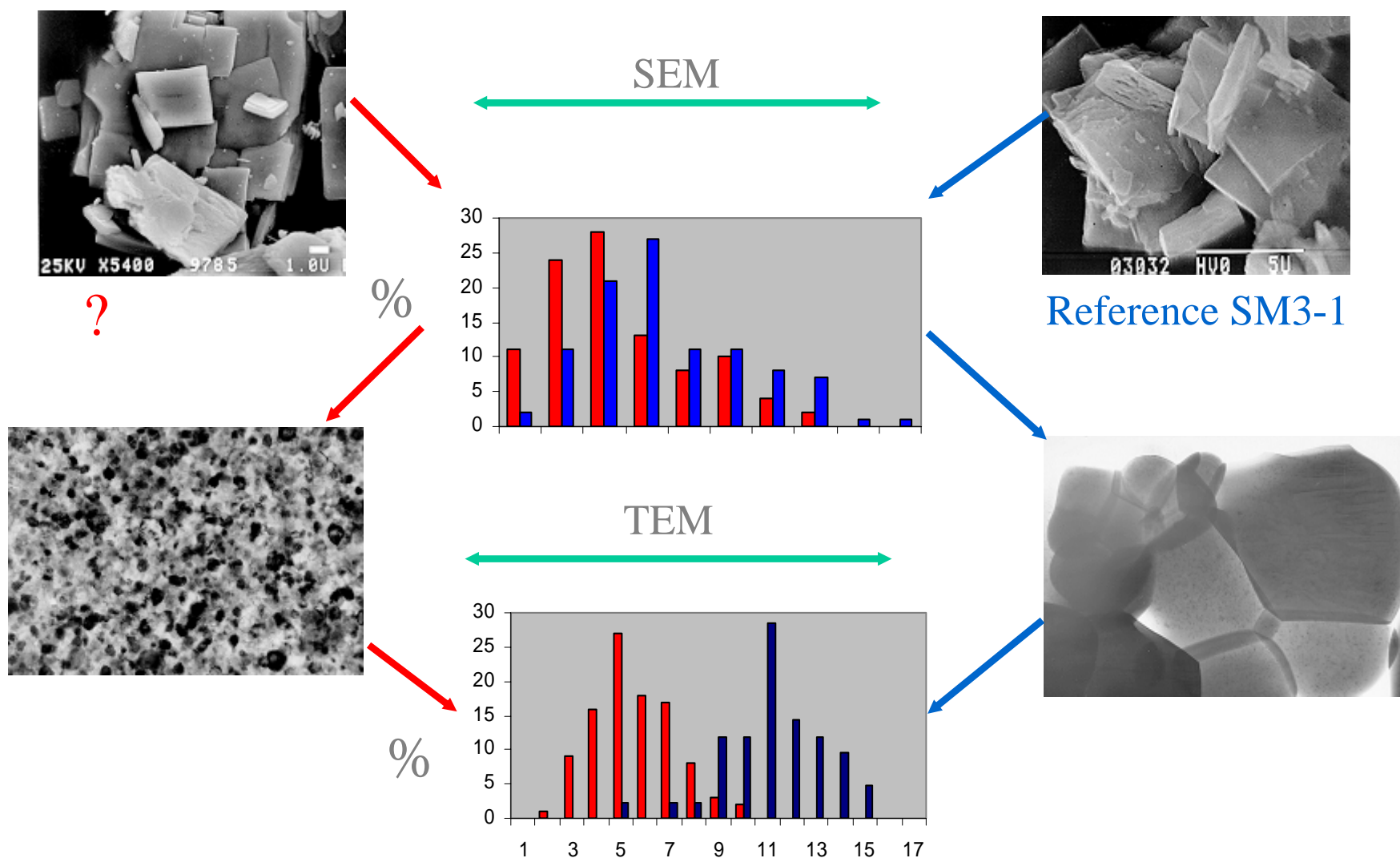
2) rod-shaped PuO_2



	Bulk	Particle 1	Particle 2
$^{240}\text{Pu}/^{239}\text{Pu}$	0.1226	0.1159	0.1245
Age from the $^{240}\text{Pu}/^{236}\text{U}^*$	(20.6)**	21.5	19.9

*Age by SIMS in February 2000. ** By gamma spectrometry from the Am-241 build-up.

Microstructural fingerprint



Plutonium

- close to weapons-grade
- not from commercial reprocessing or from military production
- mixture of residues of different types of spent fuels

Lithium-6

- energetic tritons via ${}^6\text{Li}(n,\alpha){}^3\text{T}$
- tritium can initiate DT nuclear fusion in a thermonuclear weapon

May not be a coincidence to find such materials together !!

WAK – Reprocessing plant (decommissioned)



Contamination of an employee was detected in a routine urine control.

“Players”:



Mr. M. – The thief



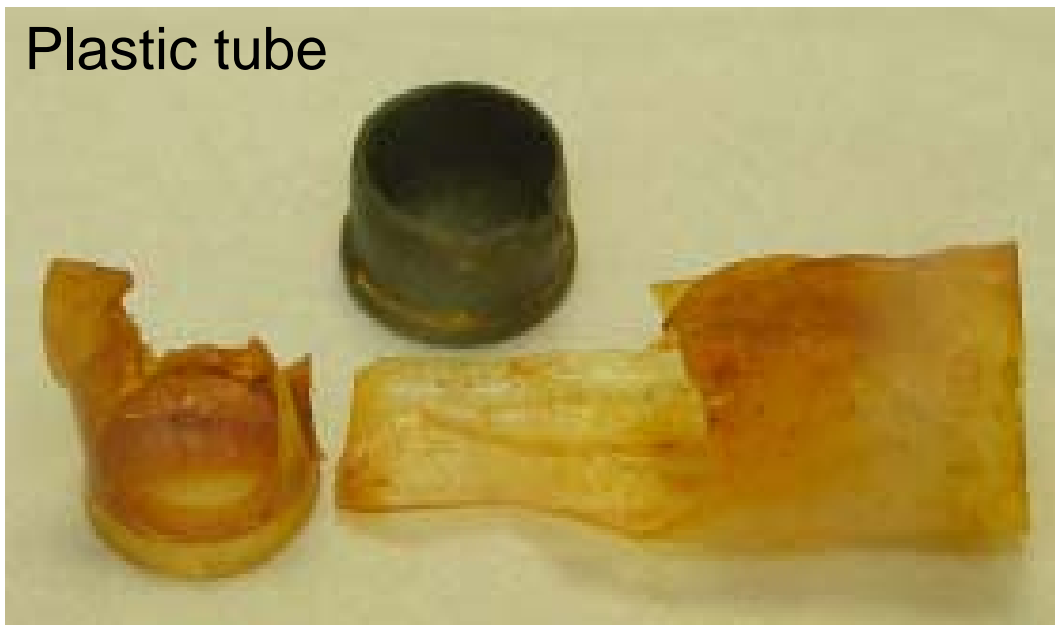
Mrs. E. - Girl friend of Mr. M.



Ms. A. E. - Daughter of Ms. E.

Stolen items:

Plastic tube



Swipe cloth



Total of a few milligrams of radioactive material including Pu, U, ^{241}Am , $^{134+137}\text{Cs}$, ^{154}Eu and ^{125}Sb .

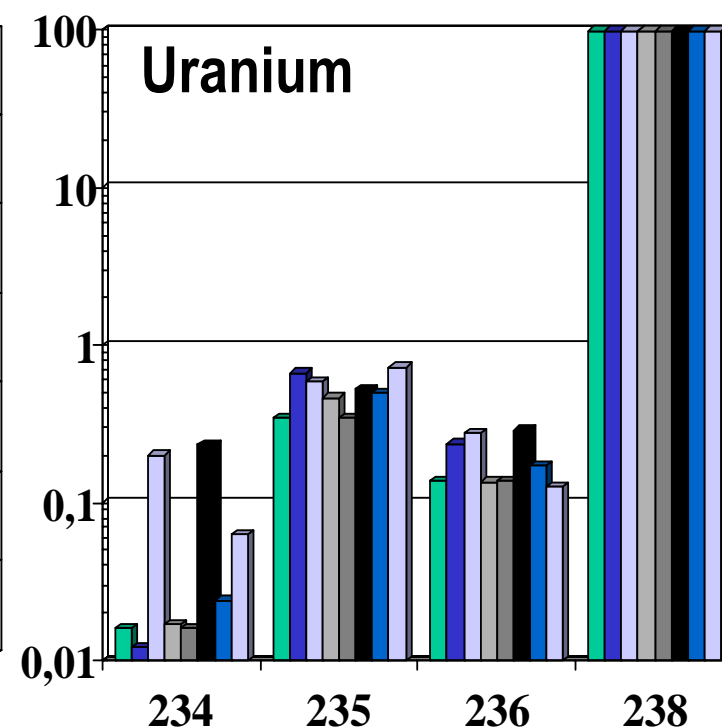
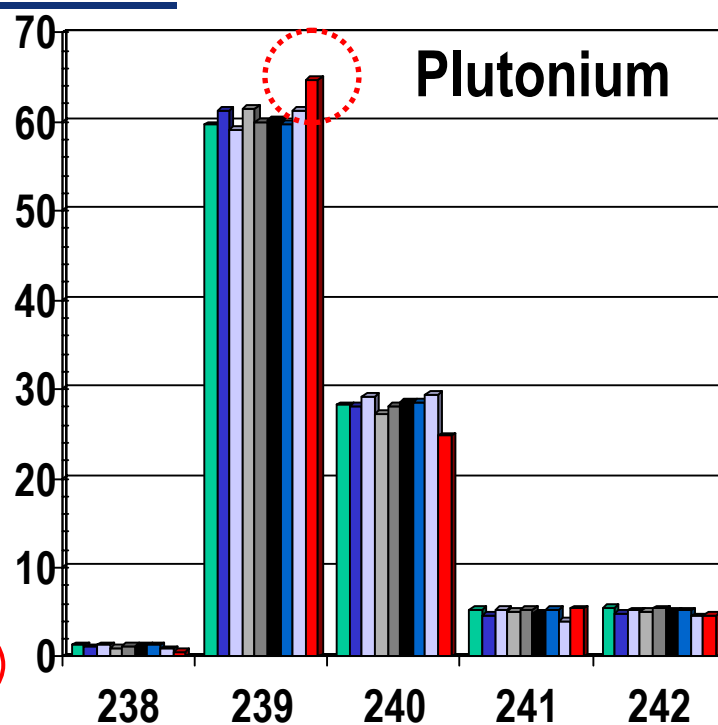
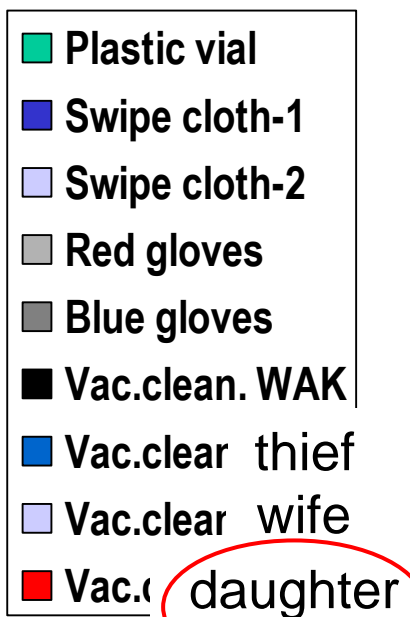
Estimated (ICRP Model) activity uptake (in kBq):

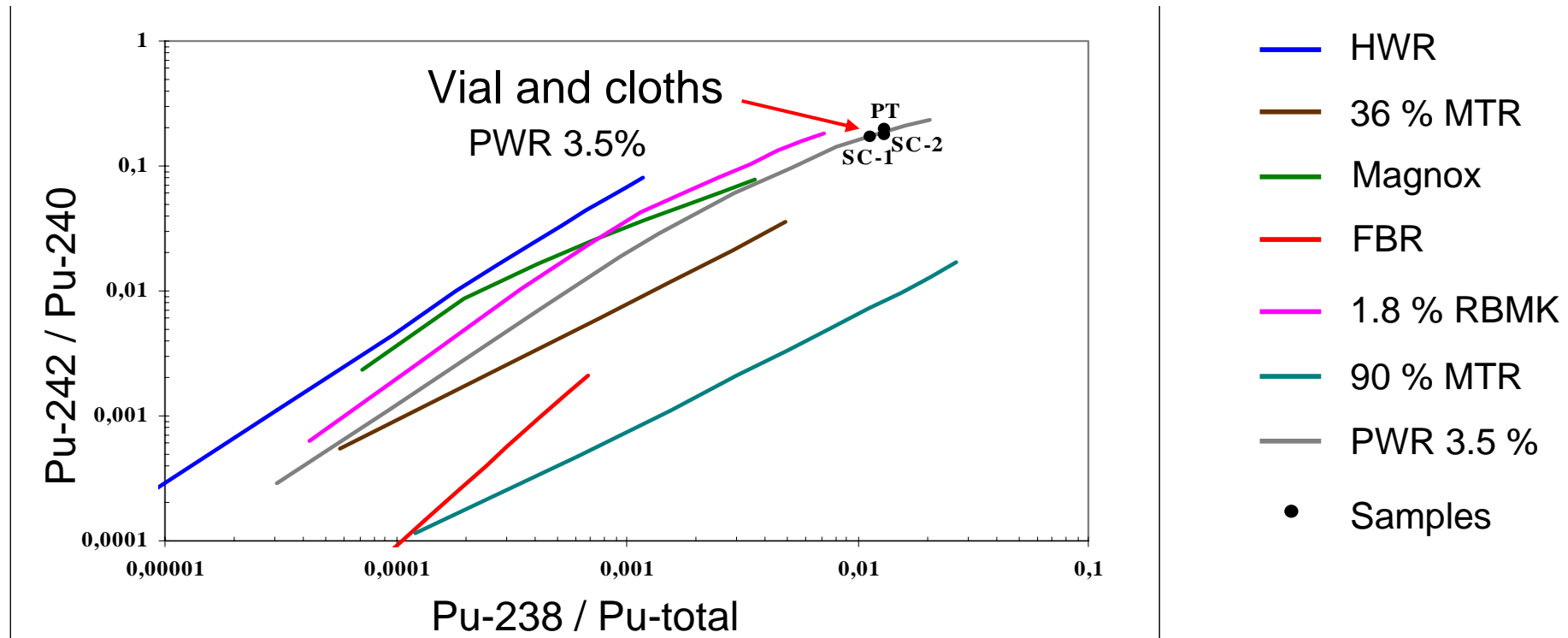
Person	Way of uptake	Cs-137	Am-241	Pu-241
Mr. M.	Inhalation		4	16
	Ingestion		-	-
Mrs. E.	Inhalation		28	1110
	Ingestion	118	3450	13800
Ms. A.E.	Inhalation		0,4	
	Ingestion	3	69	

Can the contaminations and incorporations be explained by the two sources, vial + cloth ?

- Analyse samples from different locations
 - three apartments
(thief, girl friend, daughter's boyfriend)
 - facility
- Compare
 - samples
 - external information (which fuel reprocessed, WAK, ...)

Isotopic compositions:





- Pu composition matched with the reprocessed LWR fuel
- The age of the Pu was 12-14 years, which was the time of the last reprocessing campaign

- Vacuum cleaner bag from “the daughter” showed significantly different Pu isotopic composition
 - ⇒ Pu-240/Pu-239 = 0.38 rather than 0.47
 - ⇒ mixture of reactor Pu (~ 90%) and fallout Pu (~10%)
- The plastic vial can be excluded as source of incorporation of the wife (^{137}Cs content too low)
Swipe cloth:
Varying elemental ratios can be explained by different solubilities (Cs-137 dissolves well in water, Pu and Am not)
- Decontamination of two apartments ~ 2 M€



Exercises

1. Which parent/daughter and parent/grand-daughter ratios can be used to determine the age of a 10 mg Pu-sample with the following composition (wt-%):

Pu-238: 2 %

Pu-239: 57 %

Pu-240: 25 %

Pu-241: 10 %

Pu-242: 6 %

Remarks: Method of the choice is ICP-MS without chemical separation with detection range of 10^8 (i.e. the parent/daughter ratio cannot be larger than 10^8).

2. In which of the following reactors was the Pu from previous example most likely produced ?

- BWR, 3.3 % U-235, 40 Burn-up, 8 years decay
- PWR, 3.3 % U-235, 40 Burn-up, 8 years decay
- BWR, 1.9 % U-235, 22 Burn-up, 8 years decay

Remember: $\text{Pu-240/Pu-239} = 0.44$
 $\text{Pu-241/Pu-239} = 0.18$